Table 3-18. Probe completions in the waste zone moisture monitoring array at Pit 4 and Pit 10.

	Probe Type							
	Tensiometer		Soil Moisture Probe			Tensiometer		Soil Moisture Probe	
Cluster Name	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)	Cluster Name	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)
MM1-1	MM1-1-T3	17.7	MM1-1	17.8	**MM3-2**	MM3-2-T3	8.4	MM3-2	8.5
	MM1-1-T2	10.5	MM1-1B	11.6		MM3-2-T2	6.6	MM3-2B	7.0
	MM1-1-T1	5.6	MM1-1B	5.5		MM3-2-T1	5.0	MM3-2C	4.0
MM1-2	MM1-2-T3	14.0	MM1-2	13.9	**MM3-3**	MM3-3-T3	17.0	MM3-3	17.0
	MM1-2-T2	9.3	MM1-2B	10.7		MM3-3-T1	14.0	MM3-3B	13.8
	MM1-2-T1	5.6	MM1-2B	6.0		MM3-3-T2	4.6	MM3-3B	7.5
MM1-3	MM1-3-T3	11.7	MM1-3	11.5	**MM4-1**	MM4-1-T3	18.5	MM4-1	19.4
	MM1-3-T2	8.4	MM1-3B	9.7		MM4-1-T2	14.9	MM4-1B	14.7
	MM1-3-T1	5.1	MM1-3B	4.9		MM4-1-T1	5.7	MM4-1B	6.3
MM2-1	MM2-1-T3	16.0	MM2-1	16.0				MM4-1D	16.7
	MM2-1-T2	11.9	MM2-1B	12.5	**MM4-2**	MM4-2-T3	15.8	MM4-2	17.4
	MM2-1-T1	6.7	MM2-1B	7.3		MM4-2-T2	11.4	MM4-2B	12.1
MM2-2	MM2-2-T3	9.2	MM2-2	10.8		MM4-2-T1	4.9	MM4-2B	4.7
	MM2-2-T2	8.6	MM2-2B	9.1	**MM4-3**	None installed		MM4-3	9.1
	MM2-2-T1	4.9	MM2-2B	4.0				MM4-3B	6.2
MM2-3	MM2-3-T3	6.6	MM2-3B	7.0				MM4-3C	4.8
	MM2-3-T2	5.1	MM2-3B	1.7	**MM4-4**	MM4-4-T3	9.5	MM4-4	10.3
	MM2-3-T1	3.8	MM2-3	3.0		MM4-4-T2	8.2	MM4-4B	8.7
MM3-1	MM3-1-T3	9.7	MM3-1	9.7		MM4-4-T1	3.6	MM4-4B	4.2
	MM3-1-T2	7.1	MM3-1B	7.6				MM4-4D	10.9
	MM3-1-T1	4.9	MM3-1C	4.5	**MM4-5**	MM4-5-T3	13.5	MM4-5	13.9
						MM4-5-T2	9.7	MM4-5B	9.7
						MM4-5-T1	4.1	MM4-5B	4.4

An additional array of probes, MM4, was installed to form an array around the depleted uranium focus area in the west end of Pit 10. Some probes were located in a potential topographic depression on the underlying basalt surface (Salomon 2001). Several probes in this location are located near a drainage ditch that borders the southwest corner of Pit 10. The MM4-3 cluster is located in a slight topographic depression, while the MM4-2 cluster is located in an area that has higher relative surface elevation with good surface water runoff. This location is biased toward an area of suspected low infiltration to monitor moisture behavior in an area with less favorable infiltration potential.

Additional information concerning upper basalt topography and locations where water tends to accumulate on the surface from snow melt and precipitation (e.g., ditches) was considered when selecting the locations for moisture monitoring. Upper basalt surface topography indicates a possibility for infiltrating water to move laterally toward Pits 4 and 10 from both the north and the south. Areas in the SDA that had significant ponding during a February 1995 thaw are shown in Figure 3-20 (Bishop 1996). This melting and water accumulation pattern was similar to ponding that occurred in 1993, 1994, and 1996. Water temporarily standing in ditches nearly surrounding the perimeter of Pits 4, 6, and 10 is shown in Figure 3-20. Moisture monitoring probes were placed to determine the extent of lateral movement away from these ditches into the waste zone.

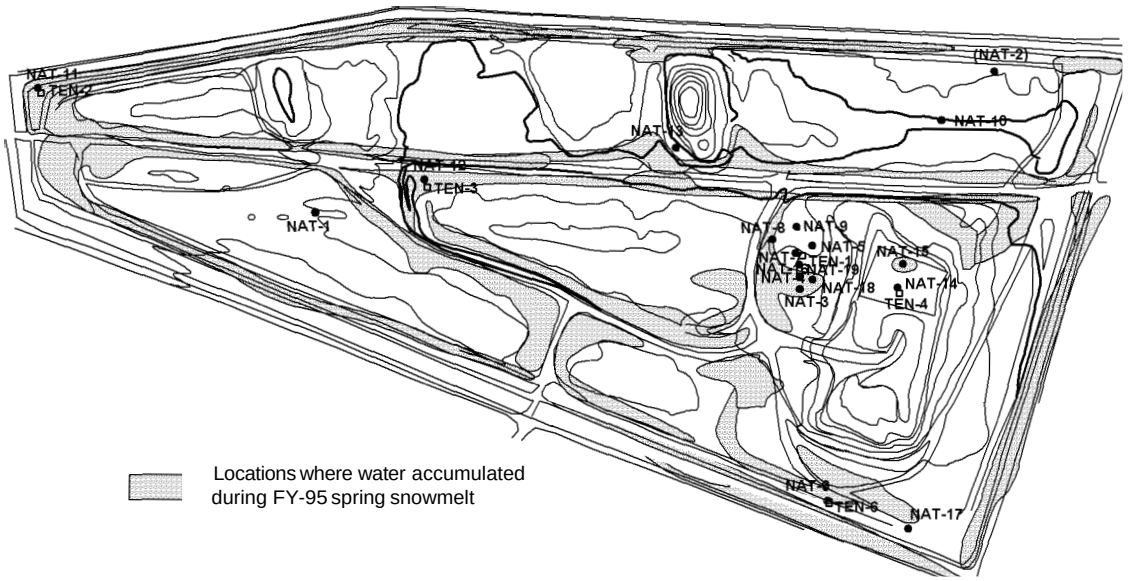


Figure 3-20. Locations at the Subsurface Disposal Area showing significant temporary accumulations of surface water during February 1995.

3.7.11 Type A Logging Results Used to Estimate Cover and Waste Zone Thickness

Results generated from nuclear logging of Type **A** probes have provided additional information to assess thicknesses of the soil cover and the waste zone. Vertical waste zone thickness and boundaries were interpreted assuming that the waste zone contains less soil, more void spaces, and increased amounts of hydrogen, iron, and chlorine compared with the overburden and underburden soil. Neutron-neutron moisture logging data were used to interpret the top and bottom of the waste zone in each Type **A** probehole, based on the assumption that low density, void spaces, and the presence of neutron absorbing elements found in typical waste can combine to cause sharp reductions in measured instrument response. Sharp decreases in the moisture tool response were first observed during Pit 9 logging in 1999. The decreased tool response occurred consistently at about the expected depth of the top of the waste zone at 1.2 to 1.8 m (4 to 6 ft) and indicated very low soil moistures of about 0 to 5% by volume below that

depth. In some cases, the moisture tool response was observed to increase sharply near the bottom of the probehole, returning to typical soil moisture values. For these reasons, the neutron-neutron tool response appears to be a valuable aid in determining vertical limits of the waste zone.

Neutron-neutron moisture logging data were the primary data set used to interpret vertical waste zone boundaries. Silicon, calcium, potassium, and thorium levels, measured using other logging tools, also were considered in cases where the moisture tool response was ambiguous. The logging methods used for the various waste zone indicators are shown in Table 3-19. Logging data from each probe were examined for sharp decreases and increases in neutron-neutron tool response. Decreases in response indicate the top of the waste and increases in tool response indicate the bottom of the waste. Depths were chosen at the approximate middle of the decrease or increase and were rounded to the nearest 0.15 m (0.5 ft). Interpreted boundaries then were compared against contamination indicators, such as gross gamma, gross neutrons, and chlorine, to ensure consistency and to recognize signal interference.

Table 3-19. Logging methods used to interpret the vertical waste boundaries.

Logging Method	Waste Zone Indication
Neutron-neutron (moisture)	Reduced response resulting from low density, void space, and neutron absorbing elements in waste
Passive spectral gamma	Reduced response resulting from reduced K-40 and Th-232 compared with native soil
Activated spectral gamma	Reduced response resulting from reduced silicon and calcium compared with native soil

Preliminary evaluations of logging data to determine overburden and waste zone thickness are summarized in Table 3-20. The waste zone thickness estimated in Table 3-20 may be greater than the value indicated because no lower waste boundary was recognized during interpretation of some logging results. In a few cases, the lower boundary of the waste zone was assumed to lie below the maximum logged depth; and, therefore, results are defined as the minimum waste zone thickness. In some logs, no indicators of waste or presence of a waste zone were indicated. Data from these probes were excluded from thickness calculations because the respective data were likely from probes installed at pit boundaries and are not representative.

Table 3-20. Overburden and waste zone thickness estimates from Type A logging data.

Pit	Number of Probes Used to Determine Estimate	Overburden Thickness (ft)	Minimum Waste Zone Thickness (ft)
Pit 4	38	7.6	11.6
Pit 5	13	5.3	7.3
Pit 9	47	5.2	5.8
Pit 10	30	6.3	8

The overburden thickness estimates listed in Table 3-20 were compared to previous evaluations conducted to estimate cover or overburden thickness. Section 3.1.5 provides information on overburden thickness summarized from Neupauer (1995) and Barnes (1989). Section 3.1.6 provides geophysical interpretation on overburden thickness estimates at selected pits within the SDA. The cover thickness estimates based on interpretations of Type A logging results correlate well with previously developed estimates.

3.7.12 Continued Data Collection in the Subsurface Disposal Area Probes

Installation of the last SDA probes was completed in November 2001 and the data collection system used to monitor the moisture monitoring array is estimated to be finished during the spring of 2002. Data have been generated from nuclear logging of Type A probes; however, interpretation of these data is still in the preliminary stages. To date, limited numbers of physical samples have been collected. Though water samples have been collected from several lysimeters, the recovered sample volume has been small, typically less than 20 mL per sample, resulting in limited analysis and high analytical detection limits. These small sample volumes are likely the result of relatively dry subsurface conditions in the SDA following installation and initial sampling. Samples collected following the 2002 spring snow melt may provide additional sample volume, which could enhance analytical performance by lowering detection levels and increasing the number of analytical parameters evaluated.

Data generated by ongoing probe sampling and monitoring may be used to validate or refute results of the modeling in this ABRA and, thus, assist in developing remedial decisions for the SDA. Data also could provide additional information useful during RD/RA. Tentative planning for OU 7-13/14 calls for continued probe data collection. Additional tasks could be considered to expand the knowledge generated by the unique monitoring capability that the probes provide. Though not an exhaustive list, the tasks below are tentatively planned for ongoing monitoring and data interpretation for the SDA probes:

- Contaminant Mass Estimate — Reevaluate existing nuclear logging data to determine the mass of total VOCs and specific radioisotopes (i.e., Am-241, Np-237, Pu-239/240, U-235, U-238, Th-228, and Th-232) remaining in the buried waste around probe clusters and to support future RD/RA.

- Waste Type Definition — Reanalyze existing nuclear logging data to evaluate each spectral peak. Current interpretation has relied on analysis of only seven to ten peaks. However, each probe spectral data file contains upwards of seventy peaks that can be used to assess the presence of different contaminants and waste types. These data would substantially improve confidence in WasteOScope and can be used to support future RD/RA.

- Moisture Movement — Evaluate tensiometer and soil moisture probe data to improve understanding of moisture infiltration through the cover soil and the SDA waste zone, validate assumptions on moisture content, define moisture gradients, assess the rates of moisture flux parameters for source release and transport models, and establish future RD/RA performance objective.

- Contaminant Migration — Evaluate data generated from lysimeters and vapor ports to obtain specific contaminant concentrations in water and vapor in the waste zone to validate transport models and establish future RD/RA performance objectives.

- Contaminant Release — Using Type A and future Type B data, evaluate the mass of contaminant in an area, in combination with the localized moisture state and contaminant concentrations. Information on the waste mass, conditions affecting moisture movement through

the system, and the contaminant concentrations could be used to validate source release modeling and provide data to establish RD/RA performance objectives.

- Waste Condition — Improve video logging techniques and review the video logs to enhance understanding of the physical nature of the waste. Subject matter experts, including personnel familiar with historical waste-generating processes and past disposal operations should be consulted to interpret the videos. This information would be used in conjunction with other data to evaluate the waste zone, increase confidence in WasteOScope, and support the development of RD/RA.

3.8 Actinide Retardation Studies

A series of experiments was conducted in laboratories at Clemson University (Fjeld, Coates, and Elzerman **2000)** to measure radionuclide retardation parameters for application to risk assessment at the SDA. The experiments focused on the actinide elements uranium, neptunium, plutonium, and americium. Experiments were conducted mainly with composite interbed sediments, with a few studies conducted with basalt. Recently, experiments have been conducted on materials collected from discrete intervals in sedimentary interbeds. Laboratory studies consisted of batch measurements, column elution experiments, and measurement of adsorption isotherms. Test solutions were made up to resemble the chemistry of groundwater near the SDA.

The objective of these experiments was to quantify the partitioning of radionuclides between vadose zone water and sedimentary interbed material from the SDA. Most experiments were designed to measure a linear, reversible partition coefficient referred to as K_d. The K_d parameter is defined as the ratio of the quantity of a contaminant sorbed onto a solid phase, such as sediment or basalt, to the quantity of the contaminant remaining in the water:

$$K_d = \frac{C_s}{C_w} \qquad \textbf{(3-1)}$$

where

K_d = linear, reversible partition coefficient (mL/g)

C_s = quantity of the contaminant on a solid (pCi/g, mg/g)

C_w = quantity of the contaminant in water (pCi/mL, mg/mL).

In column experiments, retardation of the movement of a radionuclide relative to the rate of water movement through the column was measured. The retardation of a radionuclide is related to the K_d by

$$R = 1 + \frac{\rho}{\theta} K_d \qquad \textbf{(3-2)}$$

where

R = retardation factor (unitless)

p = bulk density of sediment in the column (gram/milliliter)

θ = water content of the column (milliliter/milliliter).

Rearranging Equation (3-2), K_d can be calculated from

$$K_d = \frac{\theta}{\rho}(1 - R) \qquad (3\text{-}3)$$

Equation (3-3) is used in this section to calculate K_d values from retardation factors reported from column experiments.

Between 1993 and 1995, a series of batch adsorption experiments and a few column elution experiments were conducted at Clemson University and at the KNEEL using uranium, plutonium, and americium (Newman et al. 1996). Nine sedimentary interbed samples from multiple depths in Wells M6S and M7S, which were drilled near the RWMC, were mixed to form a composite interbed material. The composited material was sieved to a size range less than 0.25 mm. A few tests were conducted on a crushed INEEL basalt and an intact core of basalt. Tests were conducted in a synthetic groundwater with a composition similar to groundwater in the Snake River Plain Aquifer near the RWMC (pH = 8.0; HCO_3 = 96.6 mg/L). Column tests were conducted in packed columns 20 to 25 cm (8 to 10 in.) in length. Batch partition experiments were conducted following American Society for Testing and Materials (ASTM) Method D4319, "Standard Method for Distribution Ratios by the Short-Term Batch Method."

For column tests on basalt, uranium transport was reasonably well explained by a K_d type model, with about 100% recovery. Retardation, as defined in Equation (3-2), was about 2 for uranium, translating to a K_d of about 0.2 to 0.3 mL/g (see Table 3-21). Plutonium (total) and americium showed an enhanced mobility fraction of about 2% with a retardation factor, as defined in Equation (3-2), of about 2 for crushed

Table 3-21. Summary of batch and column measurements of americium, plutonium, and uranium partition coefficients from Newman et al. (1996).

Isotope	Media	Batch K_d	Column K_d	Enhanced Mobility K_d (mL/g)
Americium	Basalt	70 to 280	Greater than 60"	0.2
Americium	Interbed	450 to 1100	Greater than 48"	Present[b]
Plutonium(V)	Basalt	70 to 130		
Plutonium(V)	Interbed	5,100 to 7,900		
Plutonium(V)	Surface soil	7,800 to 22,000		
Plutonium(VI)	Basalt	12 to 24		
Plutonium(VI)	Interbed	110 to 690		
Plutonium(VI)	Surface Soil	1,800 to 4,900		
Plutonium (total)	Basalt		>60[a]	0.25
Plutonium (total)	Interbed		>48[a]	Present[b]
Uranium	Basalt	4 to 6	0.2 to 0.3	Not present
Uranium	Interbed	3 to 6	7 to 10	Not present

a. Breakthrough did not occur, and so it is only known that the K_d is greater than the reported value.

b. An enhanced mobility fraction was present, but at too small of a concentration to quantify a K_d.

basalt columns. The remaining 98% of plutonium and americium did not emerge from the crushed basalt column, indicating that retardation exceeded 200. Based on the bulk density and porosity of the column, a retardation value greater than 200 gives a K_d value greater than 60 mL/g (see Table 3-21). In columns containing packed composited interbed sediment, uranium was slightly retarded during transport through the column with 90% to 100% recovery of the tracer. Retardation ranged from 30 to 43, yielding K_d values between 7 and 10 mL/g (see Table 3-21). Plutonium and americium showed a very small fraction of enhanced transport, with more than 99% of the plutonium and americium remaining in the column indicating, a K_d greater than 48 mL/g.

A wide range of K_d values was measured for plutonium in batch adsorption experiments (see Table 3-21). For comparable materials, higher K_d values were measured when the plutonium initial oxidation state was plutonium(V), and lower when the starting oxidation state of plutonium was plutonium(VI) (see Figure 3-21). Most batch partition experiments were conducted at a water-to-solid-weight ratio of 4:1 as specified in ASTM D4319. In the vadose zone, the water-to-solid-weight ratio is closer to 1:5, as calculated with a soil bulk density of 1.5 g/cm^3 and a moisture content of 0.3 cm^3/cm^3 (Barraclough, Robertson, and Jantzer 1976), which is substantially different from the laboratory results. Oxidation state determinations on plutonium in batch experiments showed that when the solid-to-liquid ratio in experiments more closely resembled natural conditions in the vadose zone, plutonium(V) and plutonium(VI) were reduced to plutonium(IV).

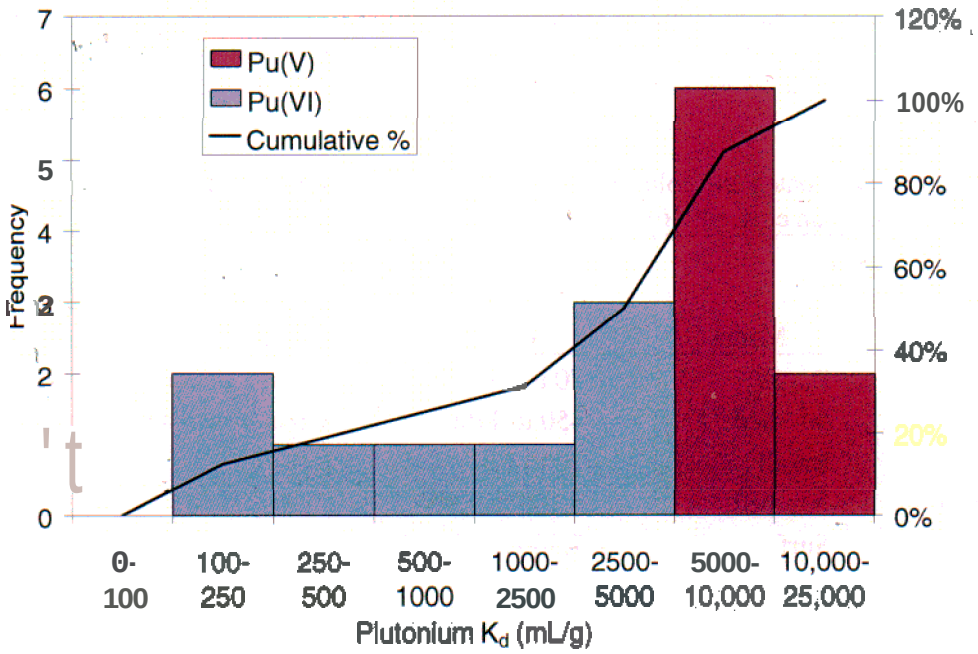


Figure 3-21, Frequency distribution of batch plutonium K_d values measured by Newman et al. (19%) differentiated by the valence state of plutonium in the starting solution.

Additional column studies were conducted at Clemson University on thorium, uranium, neptunium, plutonium, and americium to further define transport mechanisms and parameters (Fjeld, Coates, and Elzerman 2000). These studies were conducted on composite interbed material in synthetic water solution designed to resemble perched water at the RWMC (pH = 8.2; HCO_3^- = 750 mg/L). All of the experiments

were column elution experiments. This series of tests included variations in water chemistry to evaluate the effects of actinides complexing with ligands in solution.

Trivalent and quatravalent actinides, americium(III), thorium(IV), and plutonium(IV) were found to have a very small (less than 0.01%) enhanced mobility fraction with a retardation factor between 1 and 6 and a K_d less than 0.5 mL/g. Americium(III) and plutonium(IV) showed a continuous small breakthrough during the experiment. However, 99.9% of the americium(III), 98.8% of the plutonium(IV) and 99.9% of the thorium(IV) were retained in the columns with a K_d greater than 250 mL/g (see Table 3-22). Plutonium(V) showed the same behavior as plutonium(IV), with a small fraction being released from the column with little retardation, and 99.8% of the plutonium(V) remaining in the column. Plutonium(VI) was not tested.

Neptunium experiments on the column were conducted using neptunium in the neptunium(V) valence state. About half of the neptunium migrated through the columns with a retardation factor of 100 and a K_d of 25 mL/g (Table 3-22). The other half of the neptunium did not emerge from the column. Fjeld and coworkers hypothesized that some of the neptunium was being reduced from the +V to the +IV valence state in the column. A column was oxidized with hydrogen peroxide, and a breakthrough test repeated. All of the neptunium was recovered, indicating that reduction was a factor in neptunium transport. A similar experiment with plutonium(V) did not result in breakthrough of plutonium. Reduction of plutonium in the soil is strong enough that, even after treatment of the soil with hydrogen peroxide, the soil can reduce plutonium(V) to plutonium(IV).

Column experiments were conducted using the hexavalent form of uranium. Uranium(IV) can be expected to have similar geochemical characteristics as the other quatravalent actinides (Seaborg and Loveland 1990). Under reducing conditions, where uranium(IV) predominates, uranium would be strongly bound to sediments with K_d values greater than 250 mL/g, similar to values measured for thorium and plutonium(IV). Hexavalent uranium was completely recovered from the columns with a retardation factor of 3.2 to 8.2 and a K_d from 0.54 to 1.8 mL/g (Table 3-22). Experiments with water chemistry composition showed that complexing of uranium with carbonate ion was a very important factor in the transport of uranium. When carbonate ions were removed from the synthetic groundwater, the retardation factor jumped to 560 to 690 and the K_d jumped to 140 to 170 mL/g.

Complexing with organic complexing agents also was found to appreciably enhance migration of actinides. Quatravalent actinides were not mobile in the absence of complexing agents. When EDTA was added to test solutions, 27% to 61% of the quatravalent actinide became mobile with retardation factors from 16 to 79 and K_d values from 4 to 20 mL/g (see Table 3-22). Organic complexing agents were disposed of in the SDA (INEEL 1998), including unknown quantities of EDTA contained in RFP TRU waste. According to the HDT, a reasonable upper limit on the unknown quantity of EDTA from RFP is 7.1E+04 kg (LMITCO 1995a, Table 4-1). Hypothetically, the presence of these agents could enhance the mobility of actinides. However, because detections of actinides in the vadose zone are relatively sparse, the hypothesis has not been confirmed by RWMC vadose zone monitoring results (see Section 4).

During the fall of 2000, adsorption isotherms for uranium and neptunium were measured on 14 discrete interbed samples collected from boreholes drilled inside and adjacent to the SDA (Grossman et al. 2001). An adsorption isotherm is measured by a series of batch adsorption experiments carried out with different initial solution concentrations. If adsorption follows a linear adsorption isotherm (K_d), the ratio of sorbed to dissolved concentrations will be the same at all concentrations. Sediment samples from both the B-C and C-D interbeds were assessed. Experiments were again conducted in synthetic groundwater ($pH = 8.2; HCO_3^- = 220$ mg/L).

Table 3-22. Summary of retardation and K_d values measured in column experiments.

Element	Special Test Conditions	Column Retardation Factor	Column K_d (mL/g)	Fraction of Initial Spike Retained in the Column	Enhanced Mobility (Fraction Released Pore Volume)	Effective K_d of Enhanced Mobility Fraction (mL/g)
Americium		>1000	> 250"	0.999	7.9E-5	4.5
Americium	With EDTA"	16	4	0.50	7.7E-5	Continuous[b]
Thorium		>1000	>250	0.999	2.6E-5	Continuous
Thorium	With EDTA	23	5.75	0.73	2.3E-5	Continuous
Plutonium(IV)		>1000	>250	.98	6.2E-6	Continuous
Plutonium(IV)	With EDTA	35 to 79	8.8 to 20	0.42	1.7E-4	Continuous
Plutonium(V)		>1000	>250	,999	1.7E-4	Continuous
Plutonium(V)	With EDTA	>1000	>250	,999	3.6E-5	Continuous
Plutonium(V)	Oxidized sediment[d]	>1000	>250	,999	Not determined	
Neptunium		97 to 156	24 to 39	0.32 to 0.60	1.5E-4	0.3 to 0.9
Neptunium	With EDTA	58 to 80	14.5 to 20	0.33 to 0.43	1.5E-4	0.6
Neptunium	No carbonate"	195 to 310	49 to 78	0.19 to 0.33	Not present	N/A
Neptunium	Oxidized sediment	265 to 434	66 to 108	–0	Not present	N/A
Uranium		3.2 to 8.2	0.5 to 2	0.04 to 0.08	Not present	N/A
Uranium	With EDTA	Not tested				
Uranium	No carbonate	560 to 690	140 to 170	0.55	1.3E-4	3

EDTA = ethylenediaminetetraacetic acid N/A = not applicable
a. Breakthrough did not occur, and the only conclusion that can be drawn is that the K_d is greater than the reported value.
b. An enhanced mobility fraction was present, but release was more or less continuous, not as a discrete peak.
c. Simulated perched water with EDTA added.
d. Hydrogen peroxide was used to oxidize the sediment column before the test.
e. Carbonate was left out of the recipe when the simulated perched water was mixed.

For neptunium and uranium, isotherms showed distinct nonlinearity. However, a good fit for both actinides was obtained using a Freundlich isotherm (see Figure 3-22). The Freundlich isotherm is defined by the following equation:

$$C_s = K_f C_w^n \qquad (3\text{-}4)$$

where

K_f = Freundlich adsorption coefficient (mL/g)

n = Freundlich exponential coefficient (unitless).

The K values of the Freundlich isotherm showed a large amount of variation, but exponents of the isotherms n were very similar for all samples (see Table 3-23). When plotted on a log-log graph, the Freundlich isotherm becomes linear with n being the slope of the line. Parallel lines on the log-log plot indicate how similar the Freundlich n values are (see Figure 3-23).

Most of what is known about actinide transport parameters applicable to the RWMC comes from **8** years of work at Clemson University and the INEEL (Newman et al. 1996; Fjeld, Coates, and Elzerman 2000; Grossman et al. 2001). Based on these experiments, the transport of uranium and neptunium occurs in the

dissolved phase in the vadose zone, and can be represented by an adsorption type of conceptual model. Uranium, in particular, demonstrates classic breakthrough curves for column tests suggesting a reversible adsorption mechanism. Neptunium in natural waters is present in the pentavalent(V) valence state (Seaborg and Loveland 1990). *An* adsorption model for neptunium can be shown to be conservative because at least some of the neptunium is irreversibly adsorbed in column elution experiments. Experimental results from Clemson suggest that reduction of some of the neptunium(V) to neptunium(IV) causes the adsorption. Test results from Clemson indicate that adsorption isotherms for uranium and neptunium are nonlinear. As the uranium and neptunium concentration in solution increases, the partitioning ratio decreases. Sorption will not be as efficient for retarding the movement of uranium and neptunium at higher concentrations as it is at lower concentrations. A Freundlich nonlinear adsorption isotherm would, therefore, be a better approach for uranium and neptunium sorption than a constant K_d approach.

Plutonium(VI) is not stable in natural waters (Seaborg and Loveland 1990; Choppin, Bond, and Hromadka 1997; Runde 2000; Skipperud, Oughton, and Salbu 2000). Plutonium(V) is the stable oxidation state of dissolved plutonium in natural systems; however plutonium(V) is reduced to plutonium(IV) through interaction with solid minerals and soil organic matter in natural systems (Skipperud, Oughton, and Salbu 2000). Based on the findings of the Clemson batch experiments and on a review of the literature, the batch adsorption experiments at high water-to-solid ratios using plutonium(VI) as a starting material are concluded to fail to be representative of conditions expected in the subsurface at the RWMC. Some experiments where plutonium K_d values are measured using plutonium(V) as a starting material, and at low water-to-solid ratios, are more representative of the vadose zone environment. Under those conditions, a plutonium K_d over 5,000 mL/g is indicated.

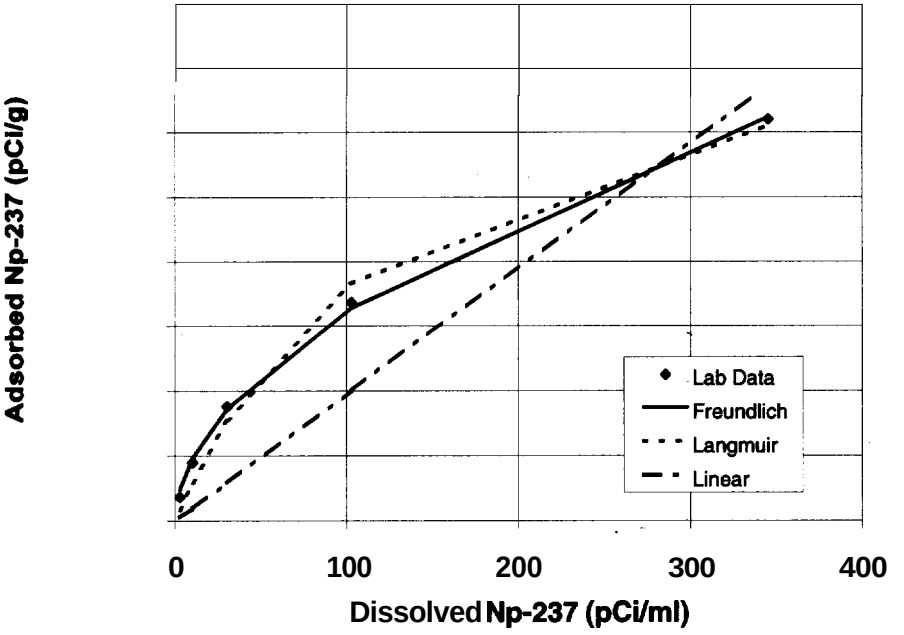


Figure 3-22. Neptunium adsorption on Sample 11s-INEEL-109 comparing the fit of Freundlich, Langmuir, and linear isotherms to the laboratory data.

Table 3-23. Measured Freundlich and linear isotherm parameters for neptunium and uranium (Grossman et al. 2001).

Sample ID	Neptunium Freundlich K	Neptunium Freundlich n	Neptunium K_d (mL/g)	Uranium Freundlich K	Uranium Freundlich n	Uranium K_d (mL/g)
7DS00101KD	246	0.59	100	74	0.81	37
7DS00301KD	91	0.62	27	53	0.78	21
7DS00901KD	100	0.59	25	17	0.78	6
7DS00701KD	24	0.65	6	46	0.76	16
7DS00501KD	155	0.56	37	37	0.81	17
7DS01701KD	133	0.56	34	40	0.77	14
7DS02301KD	109	0.56	28	35	0.81	15
I2S-INEEL-105	52	0.65	15	40	0.78	15
11s-INEEL-109	280	0.58	98	35	0.78	13
I4D-INEEL-234	86	0.54	16	23	0.76	7
I4D-INEEL-231	124	0.53	27	33	0.76	11
I1D-INEEL-234	95	0.60	25	40	0.87	22
I4D-INEEL-224	162	0.54	41	31	0.81	13
I3D-INEEL-229	256	0.57	85	27	0.77	9
Average	137	0.58	40	38	0.79	16

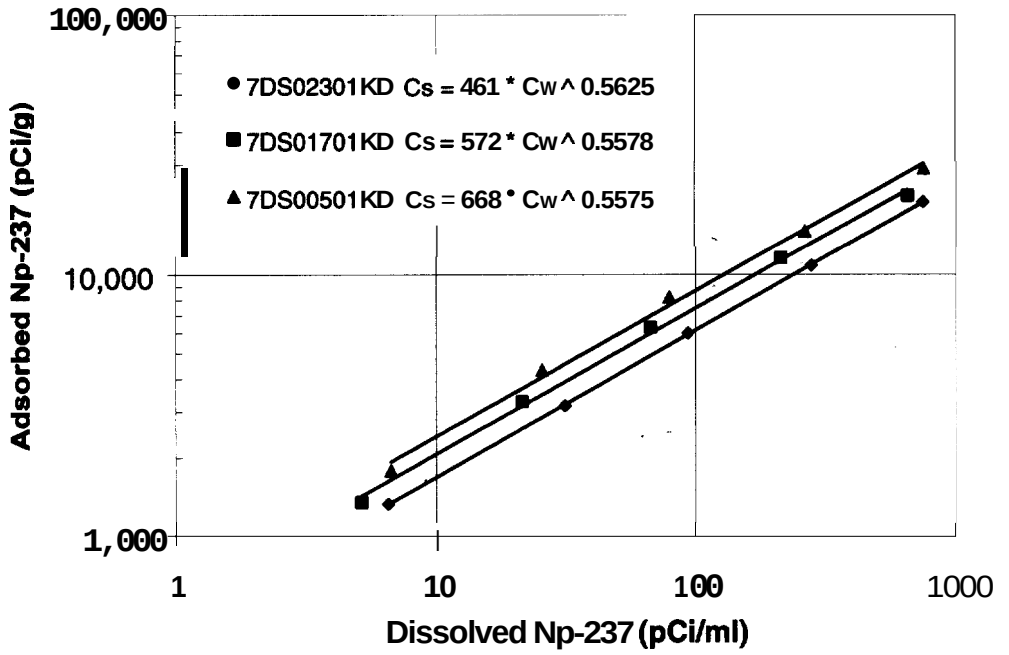


Figure 3-23. Laboratory neptunium adsorption data for three soil samples showing the similar slopes when fit with a Freundlich isotherm.

For americium and plutonium, however, transport in the dissolved phase may not be an important mechanism. Attempts to measure adsorption isotherms for americium and plutonium were unsuccessful because americium and plutonium formed insoluble mineral precipitates and could not be kept in solution in the simulated groundwater. This result is consistent with the solubility of solid plutonium and americium minerals in the synthetic water composition. Without a dissolved aqueous phase, a liquid-solid partitioning coefficient could not be measured. A better conceptual model for americium and plutonium seems to be the formation of an insoluble solid phase that is mostly filtered out of solution during batch adsorption experiments, which gives the appearance of a large Kd value in batch experiments. In column experiments, the particulate material is mostly filtered out of solution by the sediment in the column, also giving the appearance of a large Kd value for more than 98% of the plutonium.

In column experiments, however, small, but continuous release of trivalent and quatravalent actinides (thorium, plutonium, and americium) occurs, in a particulate form that migrates as fast or almost as fast as the water. This was seen in earlier column experiments conducted by Miner, Evans, and Polzer (1982) as well as in two generations of studies at Clemson University (Newman et al 1996; Fjeld, Coates, and Elzerman 2000). A continuous, slow release of a very small fraction of the actinide occurs over time. Even in the packed sediment columns where material had been sieved to sizes less than 0.25 mm, there is measurable transport of particulate actinides. Because of this characteristic, a tiny fraction of the trivalent and quatravalent actinides could be moving with little or no retardation through the vadose zone.

A final factor affecting mobility of actinides identified by the Clemson University studies (Fjeld, Coates, and Elzerman 2000) is the effect of organic complexing agents. Laboratory tests using EDTA, an organic complexing agent, showed that EDTA could greatly enhance the mobility of actinides. An inventory of complexing agents (e.g., EDTA) used at RFP (INEEL 1998) indicates that organic complexing agents are likely to be present in the waste. The quantities of EDTA from RFP are reported as unknown, but a reasonable upper limit of 7.1E+04 kg is suggested in the HDT (LMITCO 1995a, Table 4-1).

3.9 Carbon-14 Characterization

Modeling of release and migration of carbon-14 for OU 7-13/14 has been limited to dissolved-phase approaches. However, C-14 can be released and migrate as a vapor, particularly in carbon dioxide (CO_2). Recent modeling to estimate the quantities and concentrations of radioactivity in beryllium blocks, and monitoring at a beryllium disposal in SVR-20 and an activated metal disposal in SVR-12 are summarized below.

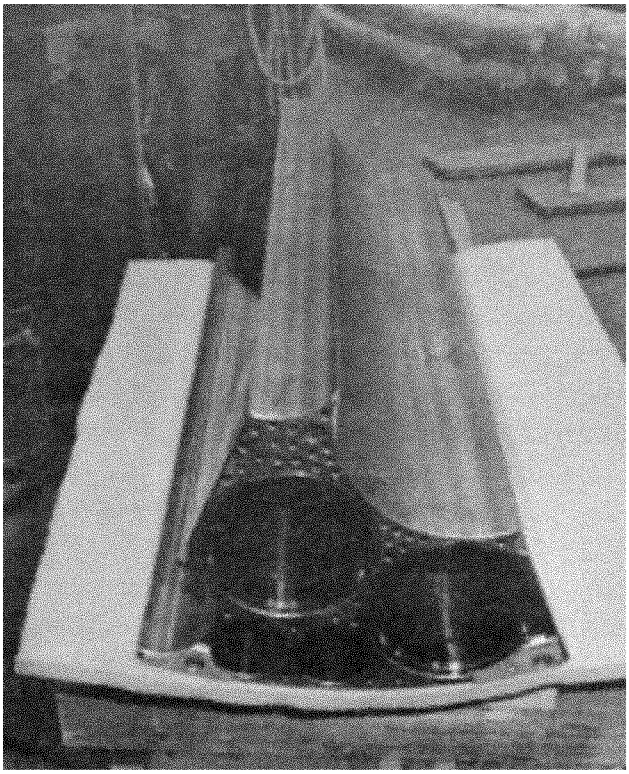
3.9.1 Estimated Activity in Beryllium Blocks Buried in the Subsurface Disposal Area

Research reactors at the INEEL TRA, including MTR, the ETR, and the ATR, use beryllium reflectors. The beryllium is used as a neutron reflector to intensify the neutron flux in the reactor core. The amount of beryllium used as a reflector varies in each reactor (Mullen et al. 2002). The ATR reflector assembly consists of a set of eight beryllium blocks and 16 outer shim control cylinders (Figure 3-24). Each block is 129.5 cm (51 in.) long, approximately 40.6 cm (16 in) square and weighs 81,420 g (179.5 lb). When all eight blocks are assembled, they have a cross section approximately 127 cm (50 in) in diameter. For the ETR, the reflector assembly was essentially four slabs that surrounded the core while the MTR reflector assembly was much more complex.

Historically, reflector assemblies from the test reactors periodically were replaced every 8 to 10 years because of swelling. The majority of irradiated beryllium reflector waste was disposed of in the SDA in three major events: 1976, 1977, and 1993. A total 5,309 kg (11,703 lb) of beryllium was disposed of from the ATR, ETR, and MTR (Table 3-24) (Mullen et al. 2002). Early characterization

efforts of the beryllium blocks relied on modeling alone to develop the isotope inventories, and there was a concern that certain key nuclides were overestimated. In an effort to reduce the conservatism in the inventory estimates samples were taken from stored irradiated beryllium blocks at TRA. The analysis was necessary to determine C-14 content. Limitations on C-14 content for LLW destined for disposal in the SDA are defined in the WAC (DOE-ID 2001) based on the LLW operation Performance Assessment (Maheras et al. 1994, Case et al. 2001). Similar analysis had not been required for previous beryllium disposals because earlier versions of the WAC did not limit C-14.

The sampling showed that the initial models were indeed conservative with regard to C-14 but substantially underestimated Nb-94 (Mullen et al. 2002). The sample results also revealed that trace impurities in the beryllium blocks, when subjected to neutron flux in a reactor, transmute to other elements. Uranium is transmuted into plutonium and other transuranic isotopes, and gold is transmuted into mercury. Modeling based on reactor burn-up histories and estimates of concentrations of impurities in the beryllium has shown that the irradiated beryllium blocks contain sufficient transuranic content to be classified as TRU waste. Table 3-24 provides estimates of the C-14 and transuranic concentrations in beryllium blocks buried in the SDA. Figure 3-25 illustrates the locations of the beryllium blocks in the SDA.



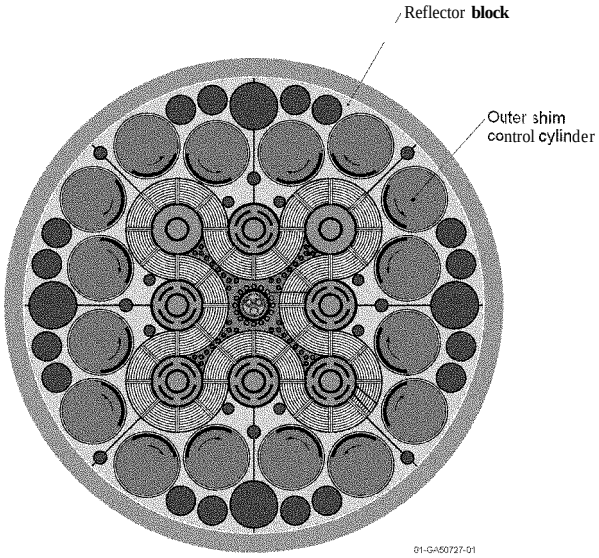


Figure 3-24. Photograph and cross-sectional view of an Advanced Test Reactor beryllium reflector block.

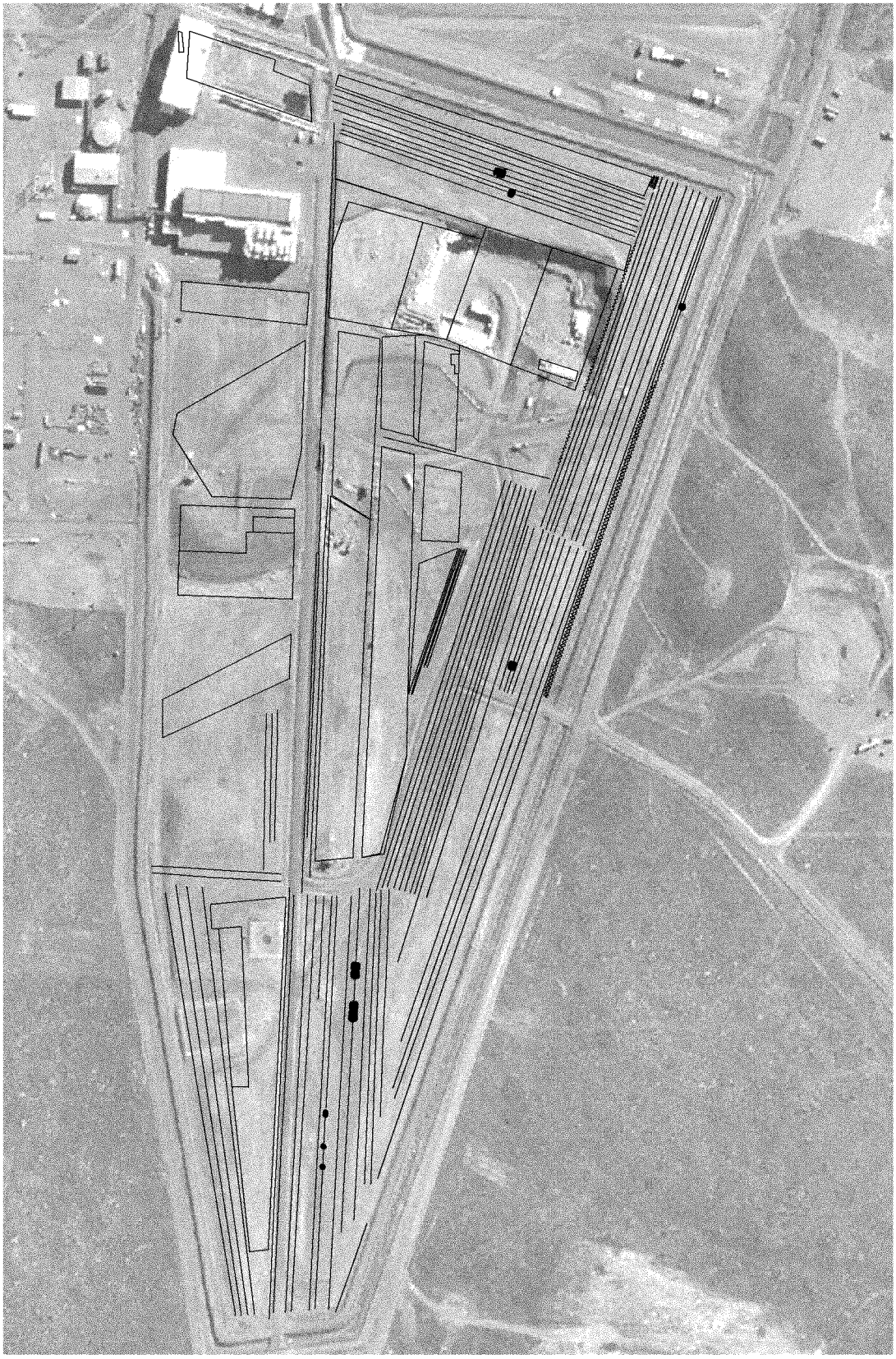


Figure 3-25. Beryllium disposal locations in the Subsurface Disposal Area

Table 3-24. Summary of the Advanced Test Reactor, Engineering Test Reactor and Materials Test Reactor transuranic and C-14 concentrations in irradiated beryllium reflector waste disposed of in the Subsurface Disposal Area as of the indicated diswosal date.

Reactor and Beryllium Waste Disposed of by Serial Number or Core Position	Initial Irradiated Date	Final Irradiated Date	Core	Reactor Position	Total Core (MWd)	Metal Mass (g)	Metal Volume (m^3)	Disposal Date	Disposal Location[a]	Total C-14 Activity (Ci)	Estimated C-14 Concentration (Ci/m^3)	Estimated Transuranic Concentration (nCi/g)	Total Transuranic Activity per Item (Ci)
Materials Test Reactor	3/3 1/52	7/1/69	1	NA	108,000	2,610,000	1.41	1977	BGT 58 3+10-20 to 3+40-50	3.78	2.68	1,000	2.61
Engineering Test Reactor	10/15/57	3/1/70	1	NA	424,3 13	580,500	0.3137	1970[b,c]	[b]	3.64	11.6	575	0.334
Advanced Test Reactor													
Block NW-L	2/1/68	9/9/72	1	NW	28,894	81,420	0.044	1976	BGT 58 200+05-15	0.9997	22.72	321.97	0.026
NW-R	2/1/68	9/9/72	1	NW	28,894	81,420	0.044	1976	Same as above	0.9997	22.72	321.97	0.026
NE-L	2/1/68	9/9/72	1	NE	27,960	81,420	0.044	1976	BGT 58 200+25-35	0.9679	22.34	321.12	0.026
NE-R	2/1/68	9/9/72	1	NE	27,960	81,420	0.044	1976	Same as above	0.9679	22.34	321.12	0.026
sw-L	2/1/68	9/9/72	1	SW	27,978	81,420	0.044	1976	Same as above	0.9683	22.01	321.13	0.026
SW-R	2/1/68	9/9/72	1	SW	27,978	81,420	0.044	1976	Same as above	0.9683	22.01	321.13	0.026
SE-L	2/1/68	9/9/72	1	SE	28,017	81,420	0.044	1976	Same as above	0.9696	22.04	321.23	0.026
SE-R	2/1/68	9/9/72	1	SE	28,017	81,420	0.044	1976	BGT 58 200+25-35	0.9696	22.04	321.23	0.026
Block NE-L	2/5/73	4/11/77	2	NE	23,625	81,420	0.044	1977	BGT 58 3+10-20	1.264	18.61	191.90	0.016
NE-R	2/5/73	4/11/77	2	NE	23,625	81,420	0.044	1977	Same as above	1.264	18.66	191.90	0.016
sw-L	2/5/73	4/11/77	2	SW	36,285	81,420	0.044	1977	Same as above	1.254	28.50	197.52	0.016
SW-R	2/5/73	4/11/77	2	SW	36,285	81,420	0.044	1977	Same as above	1.254	28.50	197.52	0.016
SE-L	2/5/73	4/11/77	2	SE	24,357	81,420	0.044	1977	Same as above	0.8441	19.18	193.52	0.016
SE-R	2/5/73	4/11/77	2	SE	24,357	81,420	0.044	1977	BGT 58 3+40-50	0.8441	19.18	193.52	0.016
Block 018L	8/9/77	2/2/86	3	NW	53,924	81,420	0.044	1993	SVR 20 0+315	1.853	42.11	387.15	0.032
013R	8/9/77	2/2/86	3	NW	53,924	81,420	0.044	1993	Same as above	1.853	42.11	387.15	0.032
015L	8/9/77	2/2/86	3	NE	47,259	81,420	0.044	1993	Same as above	1.627	36.98	406.25	0.032
019L	8/9/77	2/2/86	3	SW	60,205	81,420	0.044	1993	Same as above	2.066	46.95	370.37	0.032
014R	8/9/77	2/2/86	3	SW	60,205	81,420	0.044	1993	Same as above	2.066	46.95	370.37	0.032
011R	8/9/77	2/2/86	31	SE	72,984	81,420	0.044	1993	Same as above	2.497	56.75	340.98	0.032
Nine Outer Shim Control Cylinders	2/5/77	4/11/77	1/21	NA unknown	165,928[d]	489,881"	0.2648[f]	1987	SVR 170+10, 0+18, 1+00, 1+56	15.91	60.08	377.54	0.185
Total						5,308,781				49.8272			3.625

NA = not applicable
BGT = burial ground trench
SDA = Subsurface Disposal Area
L = left
R = right
SRV = soil vault row
TRU = transuranic
NW = northwest
NE = northeast
SW = southwest
SE = southeast
RWMC = Radioactive Waste Management Complex

a. Coordinates refer to the distance from the disposal unit boundary monument.
b. The disposal date and location at the SDA could not be determined from available records
c. The final irradiation date is assumed to be the SDA disposal date.
d. Total exposure for nine outer shim control cylinders (OSCCs).
e. Total mass for nine OSCCs.
f. Total volume for nine OSCCs.

3.9.2 Beryllium Reflector Block Monitoring

Six beryllium reflector blocks from the Advanced Test Reactor were buried in SDA Soil Vault Row (SVR)-20 in 1993 (see Figure 3-18) (fitter and McElroy 1999). The blocks contained a total of 293,000 Ci of tritiated hydrogen gas and approximately 20 of C-14. Both radionuclides form mobile compounds. About one-fifth of the total C-14 inventory in the SDA is associated with the beryllium. Carbon-14 was identified as a COPC in the IRA (Becker et al. 1998) and as a risk driver in the RWMC Performance Assessment (Case et al 2000) and the INEEL Composite Analysis (McCarthy et al. 2000). Tritium, though not a risk driver, was identified as a contaminant of interest because of its potential as a model calibration target for vapor phase transport.

Dedicated monitoring to characterize the migration of tritium and C-14 from beryllium reflector blocks buried began in 1994. Results of environmental monitoring characterizing releases and environmental conditions around the beryllium blocks in SVR-20 are summarized below.

3.9.2.1 Airborne Tritiated Water Vapor. Ambient air samples were collected 30 cm (1 ft), 1 m (3.3 ft), and 2 m (6.6 ft) aboveground near SVR-20 (see Figure 3-18) during summer and early fall of 2000 and 2001. Samples were collected weekly because the emission rate can vary substantially over relatively short periods. Passive airborne tritium samplers are used to collect the samples. Prior to 2000, samples were collected by several organizations, including the INEEL and Idaho State University for the INEEL Oversight Program. Results for all beryllium sampling are shown in Figure 3-27. Typically, the concentration of tritiated water vapor (HTO) in air ranged over several orders of magnitude each year with the peak concentrations occurring in late summer. Usually the maximum concentrations persisted over a period of a few weeks each year, but the peak concentration occurred over a period of only 4 days during 1997. The estimated annual tritium release to the atmosphere range from less than 1 Ci in 1998 to over 100 Ci in 1995 and 1996. The variation is probably caused by year-to-year differences in soil conditions that affect gas phase permeability and by possible changes in the rate of release of tritium from the blocks by corrosion. Monitoring data do not show any long-term trend in air concentration or emission, though the seasonal variability of the emission rate is evident.

3.9.2.2 Tritiated Water Vapor in Soil Gas Samples. Since 1996 soil gas samples have been collected approximately 60 to 100 cm (23 to 39 in.) from the buried beryllium blocks in SVR-20. Soil gas data through June 28, 2001, are summarized in Figures 3-28, 3-29, and 3-30. The maximum concentrations of HTO in soil moisture were 3.4, 0.28, and 0.70 μCi/mL at depths of 2.7, 4.5, and 6.2 m (9, 15, and 20 ft), respectively. Correlation of the concentrations at 4.5 and 6.2 m (15 and 20 ft) is depicted in Figure 3-31. The concentration of HTO in soil moisture appears to fluctuate over the course of each year, with lower concentrations occurring during cool months.

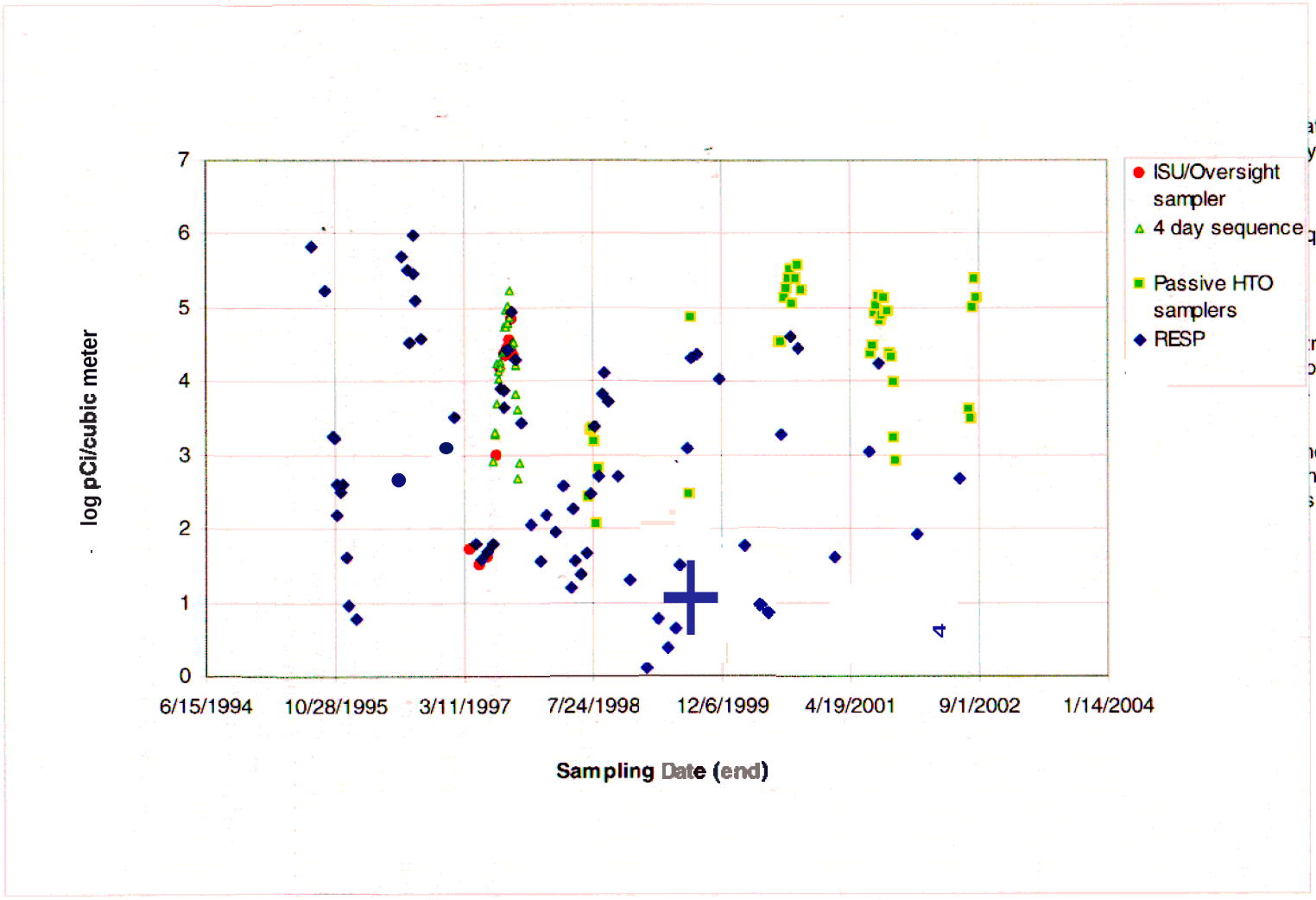


Figure 3-27. Concentrations of airborne tritiated water vapor above the beryllium blocks buried in Soil Vault Row 20.

3.9.2.3 Carbon-14 in Soil Gas. The specific activity of C-14 in CO_2 (i.e., the C-14 activity per gram of total carbon) was measured in grab samples of soil gas collected from the three GSP-1 ports and SVR20-5-VP3 at various depths. Samples collected through 1997 used a caustic solutions in bubblers and samples collected after 1997 used the Tedlar bag method . The two sampling methods are assumed to produce comparable data; however, the bag sampling and analysis procedures and calibration are still in development, and will be finalized in FY-2003. Sample results are summarized in Table 3-25. Based on a review of all results for the 1999 to 2002 monitoring, C-14 specific activity (pCi of C-14 per gram of carbon in CO_2) in soil gas near buried beryllium has increased by a factor of 5 to 10 since the late 1990s. A similar increase in HTO concentrations in soil water is evident over the same period.

Table 3-25. Summary of carbon-14 specific activity (pCi of carbon-14 per gram of carbon) in carbon dioxide in soil gas samples collected at Soil Vault Row 20.

Date	GSP-1 at 2.7 m	GSP-1 at 4.5 m	GSP-1 at 6.2 m	SVR20-5-VP3 at 5.4 m
6/5/1996	1.70E+04		2.30E+04	
7/2/1996		3.40E+04	2.50E+04	
12/12/1996	1.30E+05	4.20E+04	3.30E+04	
11/12/1997	4.40E+04	2.00E+04	1.20E+04	
11/15/2001	3.64E+05	1.58E+05	2.45E+05	8.28E+04
2/20/2002				2.85E+04
5/2/2002	1.71E+05	1.45E+05	1.33E+05	
5/23/2002				3.10E+04
8/23/2002	1.34E+05	1.33E+05	1.37E+05	3.67E+04

3.9.2.4 Chloride and Bromide in Soil Moisture. Suction lysimeters were installed at depths of 2 and 6 m (7 and 20 ft) near buried beryllium blocks in SVR-20 to collect samples of soil moisture for chemical and radiological analysis. Attempts to draw samples from the 2-m (7-ft) -deep lysimeter have been unsuccessful because of limited sample recovery. The vapor ports are either plugged or the soil surrounding the vapor ports has very low permeability. Samples were successfully collected from the 6-m (20-ft) -deep lysimeter during 1997 and 2000. Chloride was tested for in the sample analysis because magnesium chloride dust suppressant was applied on the SDA roadways in 1984 and 1985, and again in 1992 and 1993. Dissolved magnesium chloride in soil pore water would accelerate corrosion of the buried beryllium. Bromide analysis also was conducted on the 2000 sample because dust suppressant was found to have a relatively high, constant bromide-to-chloride ratio, thus being a further indicator of the impact of the dust suppressant.

The 1997 sample showed chloride concentrations in soil moisture near the beryllium blocks of about 650 ppm. This is substantially greater than the 20 to 30 ppm chloride concentrations expected in soil moisture at INEEL[i], but less than the 2,650 ppm estimate used to derive the beryllium corrosion rate for the RWMC Performance Assessment (Maheras et al. 1994). Elevated chloride concentrations

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i. Hull, Laurence, Interdepartmental personal communication with Paul Ritter, Idaho National Engineering and Environmental Laboratory, Bechtel BWXT Idaho, LLC, Idaho Falls, Idaho, March 2002.

measured around the beryllium blocks in 1997 are interpreted to be related to use of magnesium chloride dust suppressant on the SDA roads.

The chloride concentration in the 2000 sample was 35.5 ppm, and the reported bromide concentration, 0.2 ppm, was below the detection limit for the analysis (fitter and McElroy 1999). Results suggest that the chloride-bearing water present near the lysimeter in 1997 migrated downward or laterally and was replaced by water with near-background chloride concentrations.

3.9.2.5 Type B Probes. Several Type B probe vapor ports were installed at SVR-20 in 2001 to augment the three existing sampling locations at the GSP-1 borehole (Figure 3-18). Only Type B probe SVR20-5-VP3 is currently functioning. Type B Probe SVR20-5-VP3 is approximately 3.5 m (15 ft) from the centerline of the buried beryllium with vapor ports at a depth of 5.4 m. This probe is farther from the buried beryllium than the preexisting monitoring points at GSP-1 and is expected to provide usehl information concerning the lateral migration of tritium and C-14. As of March 2002, several sets of soil vapor samples have been collected for C-14 analysis as part of method development for long-term C-14 monitoring.

3.9.3 Activated Metal Monitoring

A large fraction of the total SDA inventory of C-14 is present in activated steel. The rate of release of C-14 from activated steel, accelerated by corrosion, is being studied at Soil Vault Row (SVR)-12 (Figure 3-18). Because SVR-12 is parallel to the interior perimeter road along the east edge of the SDA, it is not likely to be affected by roadway activities such as snow clearing or $MgCl_2$ dust suppressant. The area is not in an obvious runoff path, and there is little vegetation.

Highly-irradiated, activated stainless steel end pieces from spent Experimental Breeder Reactor II (EBR-II) fuel elements were disposed of in SVR-12. Spent fuel elements from EBR-II were sent to the INTEC for processing after use. The stainless steel end pieces were physically separated from the fuel in underwater basins at INTEC and sent to the SDA in 10 shipments. Personnel familiar with the subject disposals indicated that the end pieces were placed in open-top cask inserts that were perforated at the bottom to allow for draining upon removal from the storage basin (Salomon 2001).

Type B probes with vapor ports were installed near SVR-12 in 2001 (Figure 3-18). The Type B probes at SVR 12 include both vapor sampling ports and tensiometers. Tensiometers were installed to measure soil matric potential near activated metal in SVR-12 to determine hydraulic gradients, and the direction and rate of flow of soil moisture. Vapor ports were installed to provide information that could be used to characterize release and lateral migration of C-14 from the activated steel. Soil gas samples have been collected quarterly from the SVR-12 Type B probes since November 2001. In general, the specific activity of C-14 in CO_2 near the activated steel is at least 2 orders of magnitude less than the specific activity measured near activated beryllium in SVR-20. Compared to beryllium, activated stainless steel corrodes slowly and is a much less intense source of C-14.

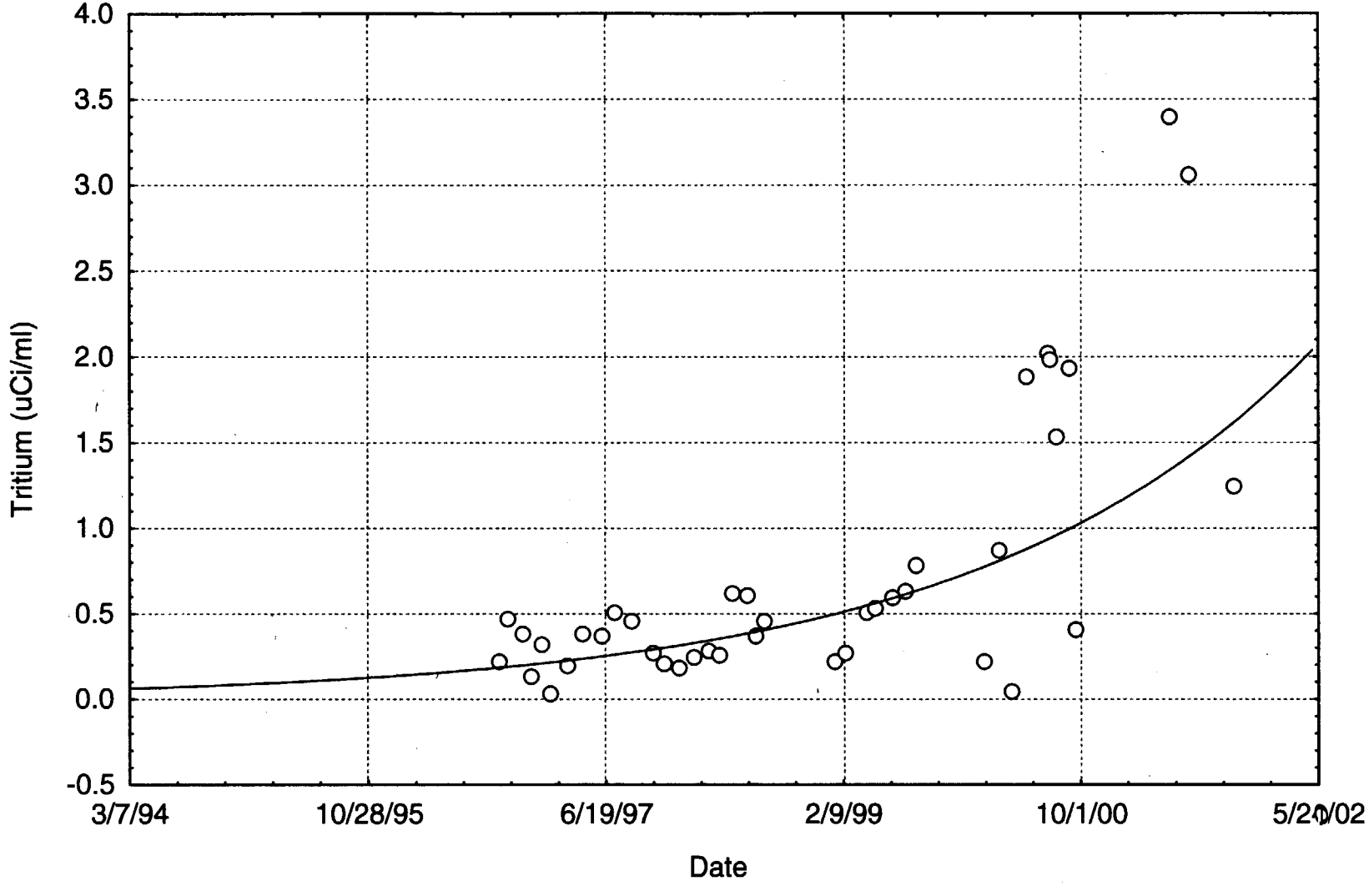

Figure 3-28. Concentrations of tritiated water in soil moisture collected from a depth of 2.7 m (9 ft) near activated beryllium in Soil Vault Row 20.

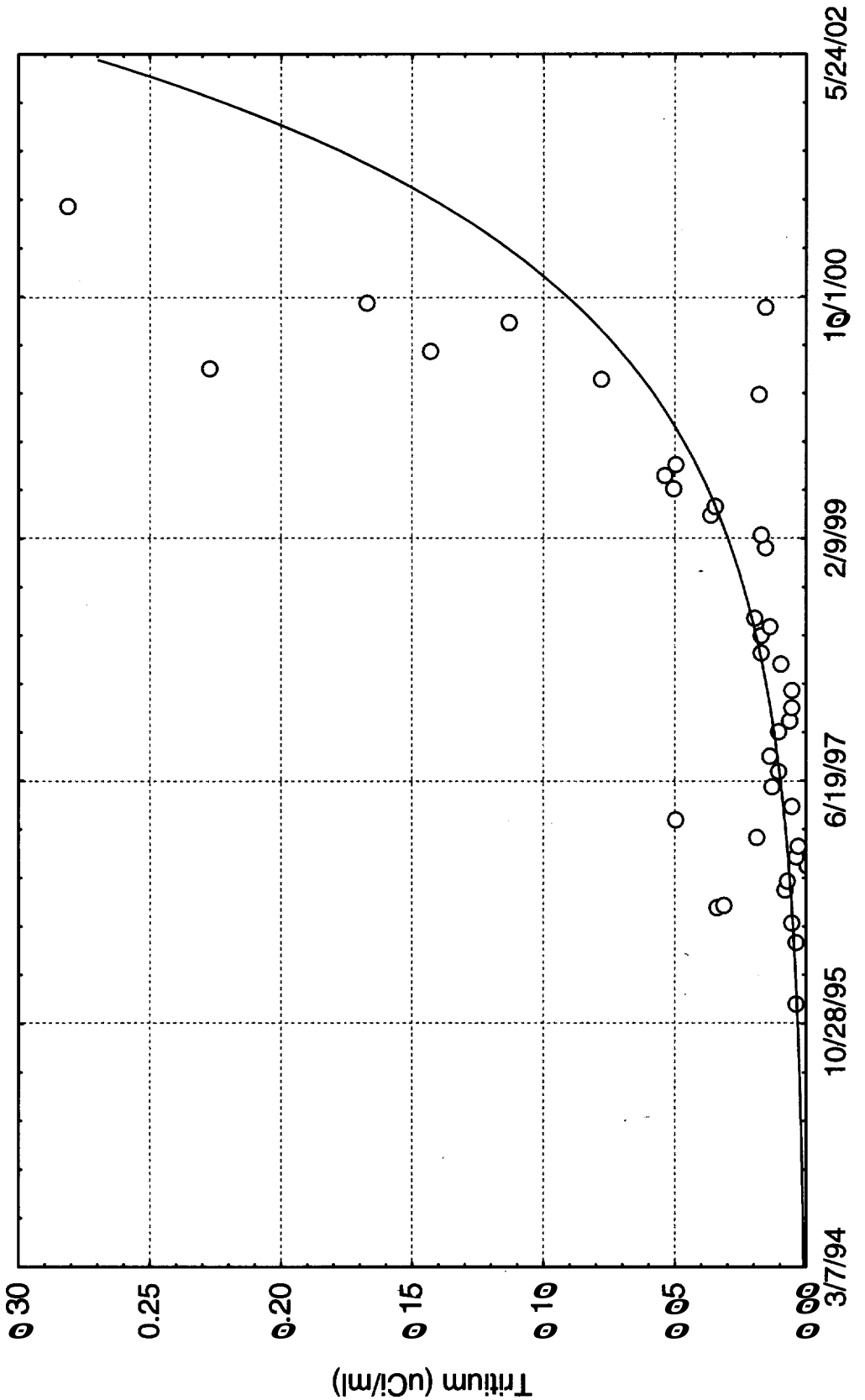

Figure 3-29. Concentration of tritiated water in soil moisture collected from a depth of 4.5 m (15 ft) near activated beryllium in Soil Vault Row 20.

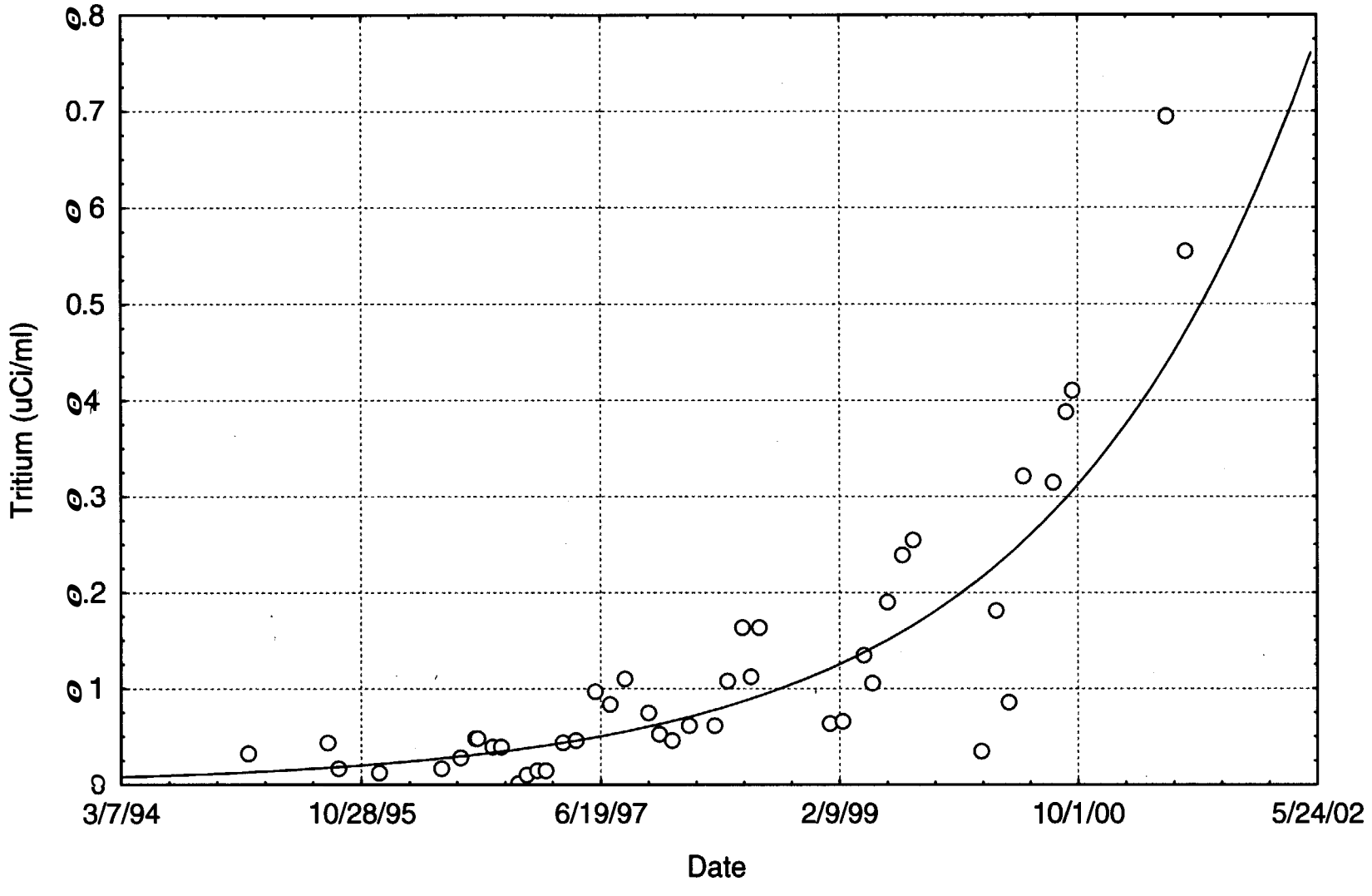

Figure 3-30. Concentration of tritated water in soil moisture collected from a depth of 6.2 m (20 ft) near activated beryllium in Soil Vault Row 20.

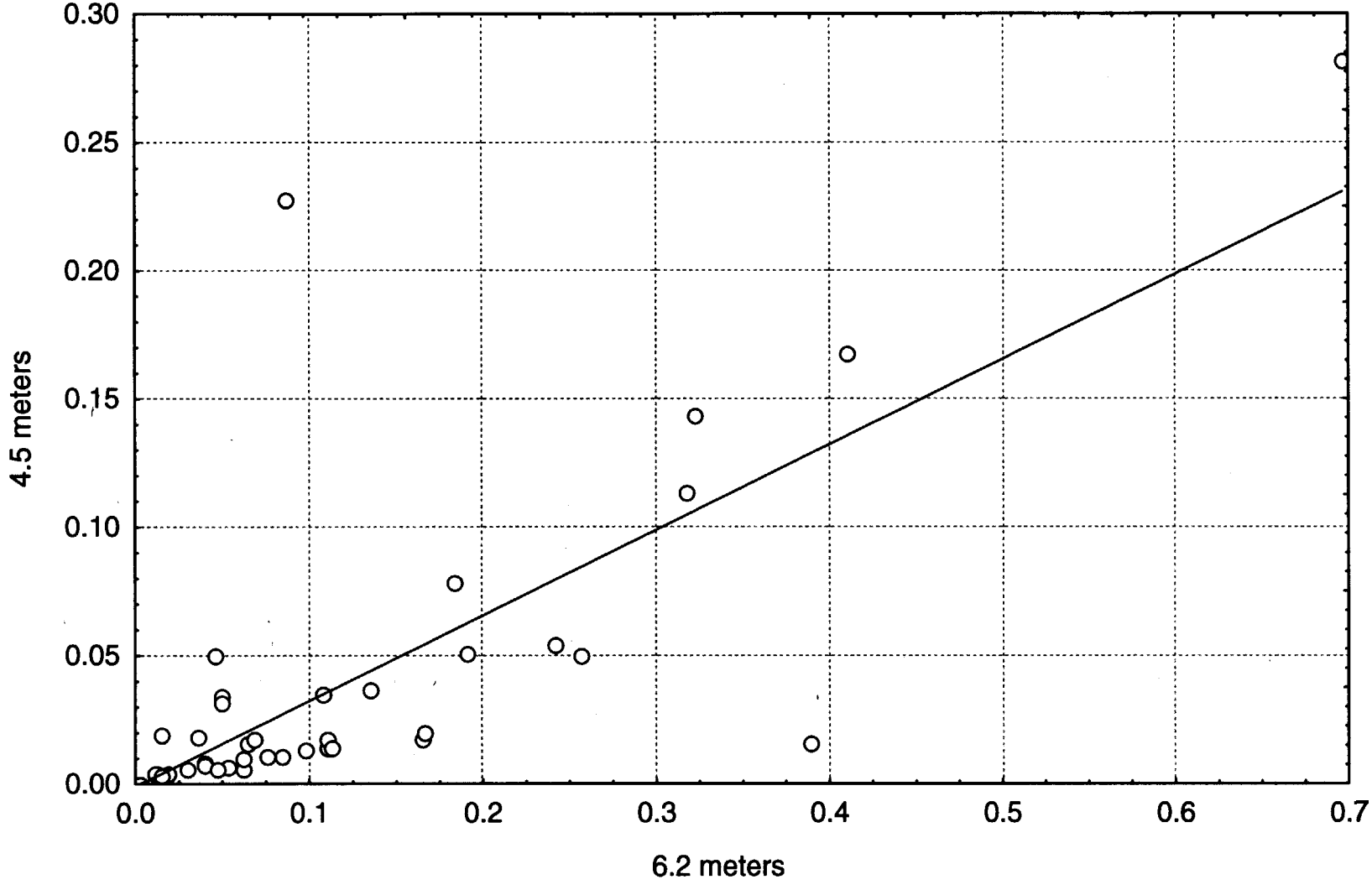

Figure 3-31. Correlation of concentrations of tritiated water in soil moisture collected from depths of 4.5 and 6.2 m (15 and 20 ft).

3.10 Preliminary Criticality Analysis for the Radioactive Waste Management Complex

The Criticality Safety Study' of the SDA was prepared to support the OU 7-13/14 comprehensive RI/FS. The study was performed in 2001 to assess postulated plutonium criticality scenarios in the SDA. The study was designed to first define waste configurations that would enhance the probability of a criticality event, then to assess sensitivity of critical configurations to various parameters affecting critical systems.

Criticality is the condition in which a nuclear fission chain reaction becomes self-sustaining. Certain isotopes will fission more readily than other isotopes do. Isotopes that tend to fission easiest are often called fissile. Plutonium is a fissile isotope that was buried in the SDA. An approximate cumulative total of 1,100 kg of Pu-238, Pu-239, and Pu-240, with an approximate combined activity of 9.9E+04 Ci was buried in the SDA. The criticality analysis was initiated because of the random manner that waste was disposed of at the SDA and the uncertainty if a criticality (a self-sustaining nuclear reaction) could occur in the buried waste.

A criticality occurs under an extremely limiting set of circumstances. Large masses of fissile material in the appropriate configuration and in the presence of a moderator (i.e., water or some other substance that slows neutrons to enhance fission) can cause a nuclear chain reaction. Parameters affecting criticality in a fissile system include (a) the mass of fissile material present, (b) the presence of moderating material, (c) the geometric configuration, (d) the presence of diluents and neutron poisons, (e) the reflection conditions around the system, and (f) the concentration and distribution of the fissile material in the waste. Each of these parameters and the effects they have on reactivity were evaluated.

Most of the plutonium waste contained in the SDA was received from RFP. No criticality concerns would exist if the buried waste complied with the fissile material disposal limits that were set at RFP. The limits were 200 g of fissile material per drum or 5 g of fissile material per cubic foot with a total loading not to exceed 350 g per box. However, SWEPP assays of stored waste at TSA indicated that some RFP drums exceed the disposal limits, or are overloaded, with respect to plutonium (Ravio et al. 1995).

Three RFP waste streams were chosen for analysis based on the perception that they could contain enough plutonium to result in a criticality in the SDA. The waste streams were identified based on limited historical SWEPP assay data from stored waste at TSA and engineering judgment, and included glovebox HEPA filters, graphite molds, and magnesium oxide waste. The HEPA filters were chosen because overloaded drums containing this waste matrix have been discovered in the aboveground storage operations at TSA. In addition, HEPA filters historically have higher fissile loading. Graphite molds were chosen based on historical data indicating the possibility of high fissile loading and on the moderation properties of graphite. Graphite is a good neutron-moderating material in large systems that consist of somewhat homogeneous distributions of fissile material. Magnesium oxide also was chosen because overloaded waste drums containing this waste matrix have been discovered in the aboveground storage operations at TSA.

Sludge waste from RFP was not considered because of the sludge waste forms and the historically low fissile loading in the sludge matrices. Most of the sludge contains a large amount of carbon tetrachloride, which is a very good neutron poison. The chlorine in the carbon tetrachloride is an excellent

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j. The study, document INEEL/EXT-01-01294, is still undergoing expert review and will be released in 2003.

neutron absorber that effectively lowers reactivity of the system by removing neutrons, thus reducing the likelihood of criticality. In addition, sludge inhibits the optimum conditions required for the formation of a critical system.

The analysis considered combinations of dumped containers and stacked containers with mass and geometry defined to enhance the probability of criticality. Regular stacking of containers would provide a more reactive geometry than randomly dumped containers, according to the analysis. Following the analysis of geometry, the amounts of moderation, neutron reflection, and neutron poison were varied to assess the potential for a configuration that could achieve criticality. The extremes of perfect neutron reflection and perfect moderation were assessed. None of the configurations analyzed was conducive to spontaneous criticality. The postulated configurations analyzed were designed to create near ideal conditions and are more reactive than the actual waste conditions within the SDA.

In summary, to achieve criticality with minimal mass, several conditions must occur simultaneously:

- The mass of plutonium must be larger than 520 g
- Water must be present in sufficient amounts to initiate a reaction
- The plutonium must be homogenously distributed within the fissile volume
- Diluting material or neutron poisons cannot be present in the system.

Clearly, within the SDA, the optimal system for criticality does not occur. The waste is intermixed with neutron poisons, such as chlorinated solvents, sludges, and other waste debris. The geometry is not optimal because the plutonium present in the waste is in isolated small masses. Substantial quantities of water necessary to moderate and sustain a reaction are not present. In addition, the metallic plutonium originally contained in the waste when it was buried would not remain in the metallic form but would transform to plutonium oxide in the environment, diminishing the likelihood of a criticality even further. Soil mixed with the waste would dilute the plutonium concentrations and the soils contain additional neutron poisons.

3.11 References

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